

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Herbert BRUNNER et al.

Serial No.: 10/574,021

Filing Date: September 24, 2004

For: High-Efficiency LED-Based Illumination System  
with Improved Color Rendering

**DECLARATION UNDER 37 C.F.R. §1.131**

We, Herbert Brunner, Tim Fiedler, Frank Jermann, and Martin Zachau, the joint inventors named in U.S. Patent Application Serial Number 10/574,021, declare as follows:

1. This declaration is submitted in support of U.S. Patent Application Serial Number 10/574,021 and, specifically to establish an invention date prior to September 23, 2003, the earliest U.S. effective filing date of published PCT Patent Publication No. WO 2004/030109 of Delsing et al., the reference cited by the Examiner in rejecting the claims under 35 U.S.C. §102(e).
2. The subject patent application is a U.S. national phase application of International application number PCT/DE04/02135 filed September 24, 2004, which claimed priority from German Patent Application No. DE 103 44 332.0 filed September 24, 2003, which is the constructive reduction to practice date of the subject invention.
3. A certified copy of German Patent Application No. DE 103 44 332.0 has been submitted to the Patent Office in connection with the present application. To further perfect the priority claim, we enclose herewith a copy of an English translation of German Patent Application No. DE 103 44 332.0 (Exhibit I).

4. We have reviewed and understand the subject application, including the claims originally filed, and any and all claim amendments made thereafter (the "Claimed Invention").

**Conception**

5. From the period prior to September 23, 2003 until the German priority Patent Application No. DE 103 44 332.0 was filed on September 24, 2003, the inventors were, in conjunction with their attorneys, diligently preparing the subject application for filing the German priority Patent Application No. DE 103 44 332.0. In fact, the German priority Patent Application No. DE 103 44 332.0 was filed September 24, 2003, which is only one day after the critical date September 23, 2003 of Delsing et al. cited by the Examiner.

6. We conceived the Claimed Invention prior to September 23, 2003 (the reference date of Delsing et al.), as evidenced by the following internal documents of OSRAM GMBH ("OSRAM"):

Exhibit 2 is a Phosphor Test Log, Attempt No. TF 119/2003 (1 page).

Exhibit 3 is an English translation of Exhibit 2.

Exhibit 4 is a Phosphor Test Log, Attempt No. TF 119/2003 (3 pages).

Exhibit 5 is an English translation of Exhibit 4.

Exhibit 6 is a graph obtained with an x-ray diffractionmeter.

Exhibit 7 is an English translation of Exhibit 6.

~~Exhibit 8 is a Notification of Invention.~~

Exhibit 9 is an English translation of Exhibit 8.

Exhibit 10 is a letter to the law firm of EPPING HERMANN FISCHER Patentanwalts-gesellschaft mbH in Munich, Germany ("EHF").

Exhibit 11 is an English translation of Exhibit 10.

These documents individually or taken together support the conception of the Claimed Invention.

**Due Diligence**

7. As shown by the date stamped on Exhibit 8, the "Notification of Invention" reflecting the Claimed Invention was submitted to and received by the IP department of OSRAM on August 12, 2003.

8. As shown in Exhibit 10, upon receiving and reviewing the "Notification of Invention", on September 5, 2003 the IP department of OSRAM forwarded the "Notification of Invention" to OSRAM's outside counsel Mr. Richard Schachtner at EHF, and requested EHF to transfer the rights from inventor Brunner to OSRAM Opto Semiconductors GmbH.

9. During the time period from September 5, 2003 to September 23, 2003, the attorneys within OSRAM GmbH studied and evaluated the subject matter described in the Notification of Invention and prepared a new patent application covering the subject matter disclosed in the Notification of Invention.

10. OSRAM GmbH subsequently filed German patent application No. DE 103 44 332.0 on September 24, 2003, to which the International application on which the subject National Phase application is based claims priority.

11. As is clear from the foregoing, the Claimed Invention was conceived prior to the September 23, 2003 reference date of Delsing et al. The inventors and those involved in preparing and filing the subject application have exhibited due diligence from August 12, 2003 up to the filing date of the German priority application on September 24, 2003.

12. We declare that all statements made herein of our own knowledge are true; that all statements made herein on information and belief are believed to be true; and further that these

statements were made with the knowledge that willful, false statements and the like are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this patent application and any patent resulting therefrom.

Date: November 23, 2009

By:

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# EXHIBIT 1

2003P14655WOUS

- 1 -

**Patent-Treuhand-Gesellschaft  
für elektrische Glühlampen mbH., Munich**

**High-efficiency LED-based illumination system with improved  
color rendering**

#### **Technical field**

- 5 The present application is closely related to the following applications:

2003P14657, 2003P14654, and 2003P14656.

- The invention is based on a high-efficiency LED-based illumination system with improved color rendering. It deals in particular with luminescence conversion LEDs which in particular are completely tunable.
- 10

#### **Discussion of background**

- One concept for a high-efficiency LED-based illumination system with improved color rendering is three-color mixing. In this concept, the mixture of the primary colors red-green-blue (RGB) is used to generate white. A blue LED can be used for partial conversion of two phosphors which emit red and green. The search for an efficient green phosphor for an RGB system is currently at the forefront of research, as is demonstrated, for example, by the proposal from US 6,255,670. Alternatively, a UV-emitting LED which excites three phosphors respectively emitting in the red, green and blue is used, cf. WO 97/48138. Examples include line emitters, such as YOB:Ce, Tb (green) and YOS:Eu (red). This requires a relatively short-wave emission (UV region < 370 nm) to enable high quantum efficiencies to be
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2003P14655WOUS

- 2 -

achieved. This requires the use of sapphire substrates for the UV-LED, which are very expensive. On the other hand, if a UV-LED based on the less expensive SiC substrates is used, one has to be satisfied with an emission in the range from 380 to 420 nm. The individual colors of the RGB system can in principle be generated by the primary radiation of LEDs or by luminescence conversion LEDs, as is illustrated by WO 01/41215.

To increase the overall light yield, a complicated system including a fourth LED which emits in the range from 575 to 605 has also been proposed in WO 00/19141. By its very nature, a system of this type is significantly more intricate, expensive and complicated than an RGB system.

DE-1 A 101 37 042 shows a slightly different concept, proposing a planar illumination system with special introduction of the blue component in order to avoid the usual absorption problems encountered with blue phosphors.

One interesting class of phosphors for illumination systems of this type are those of the oxynitridosilicate type, as are known per se under the shortened formula MSiON; cf. for example "On new rare-earth doped M-Si-Al-O-N materials", J. van Krevel, TU Eindhoven 2000, ISBN 90-386-2711-4, Chapter 6. They are doped with Tb. Emission is achieved under excitation by 365 nm or 254 nm.

A new type of phosphor is known from the as yet unpublished EP patent application 02 021 117.8 (Docket 2002P15736). It consists of Eu- or Eu,Mn-coactivated oxynitridosilicate of formula  $MSi_2O_2N_2$  ( $M = Ca, Sr, Ba$ ).

2003P14655WOUS

- 3 -

**Summary of the invention**

It is an object of the present invention to provide an LED-based illumination system with improved color rendering in accordance with the preamble of claim 1 with a color rendering which is as high as possible. A further object is to provide a tunable illumination system.

This object is achieved by the characterizing features of claim 1. Particularly advantageous configurations are to be found in the dependent claims.

The use of RGB-LEDs, which comprise three chips with the emission colors RGB, is of interest for certain applications. Since all three colors are realized by different LEDs, it is possible for all three components to be actuated independently of one another. Therefore, with this type of illumination system it is possible to deliberately set virtually any desired color locus by means of corresponding control electronics. One drawback of this solution is a very low color rendering index  $R_a < 50$ , which results from the narrow-band nature of the three individual emissions. A further drawback is that the green LED used, for technological reasons, is significantly less efficient than the other two components. An additional factor is that the color locus is highly dependent on the operating current and the temperature. Current technology (InGaN LED for blue 430 to 470 nm and InGaAlP LED for yellow  $> 540$  nm, in particular red in the range from 600 to 700 nm) has not provided a convincing solution for the primary radiation of an LED to be in the green spectral region. However, the advantage of the RGB solution realized using primary radiation is that illumination systems of this type are tunable.



2003P14655WOUS

- 4 -

On the other hand, for illumination systems in which high demands on the color rendering are to the fore, LEDs in which some of the primary LED emission is converted into light with a longer wavelength, in particular green, are used. However, such  
5 a structure is not tunable, since the secondary component is not dependent on the primary component.

Hitherto, there has not been a high-efficiency green-emitting phosphor which is at the same time insensitive to external influences.

10 The illumination system according to the invention makes simultaneous use of the color-mixing principle of blue, green and red (RGB mixing) and the principle of converting a primary radiation emitted by an LED into light with a longer wavelength by means of a phosphor which absorbs this radiation, at least  
15 two LEDs being used, of which a first LED emits primarily in the range from 420 to 470 nm (dominant wavelength) and a second LED emits in the red region at 600 to 700 nm (dominant wavelength), wherein the green component is produced by the primary radiation of the first LED being at least partially  
20 converted by a green-emitting phosphor, the green-emitting phosphor used being a phosphor from the class of the oxynitridosilicates, having a cation M and the empirical formula  $M_{(1-c)}Si_2O_2N_2:D_c$ , M comprising Sr as a constituent and D being doped with divalent europium, with  $M = Sr$ , or  $M = Sr_{(1-x-y)}Ba_yCa_x$  with  $x+y < 0.5$  being used, the oxynitridosilicate  
25 completely or predominantly comprising the high-temperature-stable modification HT.

The invention proposes a phosphor which represents an oxynitridosilicate of formula  $MSi_2O_2N_2$  ( $M = Ca, Sr, Ba$ ) which is  
30 activated with divalent Eu, if appropriate with the further addition of Mn as co-activator, with the HT phase forming the

2003P14655WOUS

- 5 -

majority or all of the phosphor, i.e. more than 50% of the phosphor. This HT modification is distinguished by the fact that it can be excited within a broad band, that it is extremely stable with respect to external influences, i.e. does not reveal any measurable degradation at 150°C, and that it has an extremely good color locus stability under fluctuating conditions (little drift detectable between 20 and 100°C). Further plus points include its low absorption in the red, which is particularly advantageous in the case of phosphor mixtures. This phosphor is often also referred to below as Sr Sion:Eu.

When producing the novel phosphor, it is important in particular to use a high temperature, the synthesis range lying between 1300 and 1600°C. Another determining factor is the reactivity of the starting components, which should be as high as possible.

The phosphor  $\text{MSi}_2\text{O}_2\text{N}_2:\text{Eu}$  ( $\text{M} = \text{Ca}, \text{Sr}, \text{Ba}$ ) which is known from EP patent application 02 021 117.8, in the case of the Sr-dominated embodiment with  $\text{M} = \text{Sr}$  or  $\text{M} = \text{Sr}_{(1-x-y)}\text{Ba}_y\text{Ca}_x$  with  $x + y < 0.5$ , referred to below as Sr Sion, is difficult to control. Although some tests give excellent results, there has hitherto been no guiding principle as to how to obtain reliable and reproducible results. An additional factor is a certain tendency of the efficiency to be reduced and the color locus to vary under high thermal loads.

Surprisingly, it has now been found that the two phases fundamentally differ in terms of their suitability for use as a phosphor. Whereas the LT phase is virtually of no use as an Eu-doped phosphor and only emits weak orange-red light, the HT phase has an excellent suitability for use as a phosphor which

2003P14655WOUS

- 6 -

emits green light. There is usually a mixture which manifests both forms of emission over a broad band. It is therefore crucial for the HT phase to be produced in as pure a form as possible, in a proportion of at least 50%, preferably at least 70%, particularly preferably at least 85%.

- This requires an annealing process which is carried out at at least 1300°C but no more than 1600°C. A temperature range from approximately 1450 to 1580°C is preferred, since LT phase is formed to an increasing extent at lower temperatures and the phosphor becomes increasingly difficult to process at higher temperatures; above approximately 1600°C it forms a hard-sintered ceramic or melt. The optimum temperature range depends on the precise composition and the properties of the starting materials.
- 15 A batch of the starting products which is substantially stoichiometric using the base components  $\text{SiO}_2$ ,  $\text{SrCO}_3$ , and  $\text{Si}_3\text{N}_4$  is particularly important for producing an efficient phosphor of the Sr Sion type. Sr is representative of M in this context. The deviation should amount to no more than in particular 10%, preferably 5%, from the ideal stoichiometric batch, including any addition of a melting auxiliary, as is often customary. A maximum deviation of 1% is particularly preferred. In addition, there is a europium fraction for the doping, realized, for example, as oxide  $\text{Eu}_2\text{O}_3$ . This discovery runs contrary to the previous procedure of adding the base component  $\text{SiO}_2$  in a significantly substoichiometric proportion. This discovery is also particularly surprising on account of the fact that other Sions which are recommended for use as phosphors, such as Ba Sion in accordance with the teaching of EP patent application 02 021 117.8, should indeed be produced with a substoichiometric quantity of  $\text{SiO}_2$ .

2003P14655WOUS

- 7 -

Therefore, a corresponding batch for the Sr Sion  $\text{MSi}_2\text{O}_2\text{N}_2$  uses 11 to 13% by weight of  $\text{SiO}_2$ , 27 to 29% by weight of  $\text{Si}_3\text{N}_4$ , remainder  $\text{SrCO}_3$ . Ba and Ca fractions in M are correspondingly added as carbonates. Europium is added, in accordance with the desired doping, for example as an oxide or fluoride, as a replacement for  $\text{SrCO}_3$ . The batch  $\text{MSi}_2\text{O}_2\text{N}_2$  is also to be understood as encompassing any deviations from the exact stoichiometry, provided that they are compensated for with a view to charge retention.

It has proven particularly expedient for the starting components of the host lattice, in particular  $\text{Si}_3\text{N}_4$ , to have the highest possible purity. Therefore,  $\text{Si}_3\text{N}_4$  which is synthesized from the liquid phase, for example starting from silicon tetrachloride, is particularly preferred. In particular the contamination with tungsten, cobalt, aluminum and calcium has proven critical. The impurity levels should in each case be less than 100 ppm, in particular less than 50 ppm. Furthermore, the highest possible reactivity is advantageous; this parameter can be quantified by the reactive surface area (BET), which should be at least  $6 \text{ m}^2/\text{g}$ , advantageously at least  $8 \text{ m}^2/\text{g}$ .

In the event of a deviation from the above procedure with regard to stoichiometry and temperature, increasing levels of undesirable foreign phases, namely nitridosilicates  $\text{MxSi}_y\text{N}_z$ , such as for example  $\text{M}_2\text{Si}_5\text{N}_8$ , are formed if the addition of  $\text{SiO}_2$  is set at too low a level, so that an excess of nitrogen is produced. Although this compound per se is a useful phosphor, with regard to the synthesis of the Sr Sion, it is extremely disruptive just like other nitridosilicates, since these foreign phases absorb the green radiation of the Sr Sion and may convert it into the known red radiation provided by the

2003P14655WOUS

- 8 -

nitridosilicates. Conversely, if too much  $\text{SiO}_2$  is added, Sr silicates, such as for example  $\text{Sr}_2\text{SiO}_4$ , are formed, since an excess of oxygen is produced. Both foreign phases absorb the useful green emission or at least lead to lattice defects such as vacancies, which have a considerable adverse effect on the efficiency of the phosphor. The starting point used is the basic principle that the level of the foreign phases should be below 15%, preferably even below 5%. In the XRD spectrum of the synthesized phosphor, this corresponds to the requirement that with the XRD diffraction angle  $2\theta$  in the range from  $25^\circ$  to  $32^\circ$ , the intensity of all the foreign phase peaks should be less than  $1/3$ , preferably less than  $1/4$ , particularly preferably less than  $1/5$ , of the intensity of the main peak characterizing the HT modification at approximately  $31.8^\circ$ . This applies in particular to the foreign phases of type  $\text{Sr}_x\text{Si}_y\text{N}_z$ , in particular  $\text{Sr}_2\text{Si}_5\text{N}_8$ .

With an optimized procedure, it is reliably possible to achieve a quantum efficiency of from 80 to well over 90%. By contrast, if the procedure is not specific, the efficiency will typically lie in the range from at most 50 to 60% quantum efficiency.

Therefore, according to the invention it is possible to produce a phosphor which represents an oxynitridosilicate of formula  $\text{MSi}_2\text{O}_2\text{N}_2$  ( $\text{M} = \text{Ca}, \text{Sr}, \text{Ba}$ ) which is activated with divalent Eu, if appropriate with the further addition of Mn as co-activator, with the HT phase forming the majority or all of the phosphor, i.e. more than 50% of the phosphor. This HT modification is distinguished by the fact that it can be excited within a broad band, namely in a wide range from 250 to 480 nm, that it is extremely stable with respect to external influences, i.e. does not reveal any measurable degradation at  $150^\circ\text{C}$  in air, that it has an extremely good color locus stability under fluctuating

2003P14655WOUS

- 9 -

conditions. Further plus points include its low absorption in the red, which is particularly advantageous in the case of phosphor mixtures. This phosphor is often also referred to below as Sr SiO<sub>3</sub>:Eu. A majority of the HT modification can be

5 recognized, inter alia, from the fact that the characterizing peak of the LT modification in the XRD spectrum at approximately 28.2° has an intensity of less than 1:1, preferably less than 1:2, compared to the peak with the highest intensity from the group of three reflections of the HT

10 modification which lie in the XRD spectrum at 25 to 27°. The XRD spectra cited here in each case relate to excitation by the known Cu-K<sub>α</sub> line.

With the same activator concentration, this phosphor reveals different emission characteristics than the LT variant of the

15 same stoichiometry. The full width at half maximum of the HT variant is significantly lower in the case of the optimized HT variant than in the case of the mixture containing foreign phases and defects or high levels of foreign phases, and is in the range from 70 to 80 nm, whereas the specimen containing

20 foreign phases and defects is at approximately 110 to 120 nm. The dominant wavelength is generally shorter, typically 10 to 20 nm, in the case of the HT modification than in the case of the variant containing significant levels of foreign phases. An additional factor is that the efficiency of the high-purity HT

25 modification is typically at least 20% higher, and in some cases significantly higher still, than in the case of the LT-dominated mixture or the mixture with a high level of foreign phases.

One characterizing feature of a sufficiently low level of the

30 NT modification and foreign phases is a full width at half maximum (FWHM) of the emission of less than 90 nm, since the

2003P14655WOUS

- 10 -

lower the level of foreign phases, the lower the proportion of the specific orange-red emission from the modification which is rich in foreign phases, in particular the nitridosilicate foreign phases Sr-Si-N-Eu in particular Sr<sub>2</sub>Si<sub>5</sub>N<sub>8</sub>:Eu.

- 5 The abovementioned typical reflections in the XRD spectrum, which reveal the different crystal structure, are another important factor, in addition to the reduced full width at half maximum.

- 10 The dominant peak in the XRD spectrum of the HT modification is the peak at approximately 31.7°. Other prominent peaks are the three peaks of approximately the same intensity between 25 and 27° (25.3 and 26.0 and 26.3°), with the peak with the lowest diffraction being the most intensive. A further intensive peak is 12.6°.

- 15 This phosphor emits predominantly green light with a dominant wavelength in the range 555 to 565 nm.

It is also possible to add a small amount of AlO instead of SiN (in an amount of up to at most 30% of the SiN content).

- 20 Both phases of the Sr Si<sub>2</sub>N<sub>2</sub>:Eu can crystallize analogously to the two structurally different host lattice modifications and can each be produced using the SrSi<sub>2</sub>O<sub>2</sub>N<sub>2</sub>:Eu batch stoichiometry. Minor deviations from this stoichiometry are possible. The Eu-doped host lattices surprisingly both luminesce when excited in the blue or UV region, but in each
- 25 case after host lattice modification with a different emission color. The LT modification reveals an orange emission, the HT modification a green emission at approximately  $\lambda_{\text{dom}} = 560$  nm with in principle a significantly higher efficiency. A desired property of the phosphor can be set accurately as a function of

2003P14655WOUS

- 11 -

the dopant content and dopant material (Eu or Eu, Mn) and the relative proportions of the HT and LT modifications.

One benefit of the HT phase is the fact that it can be excited with a good level of uniformity over a very wide spectral  
5 region with only minor variations in the quantum efficiency.

Moreover, within a wide temperature range the luminescence of the HT modification is only weakly dependent on the temperature. Therefore, the invention has for the first time discovered a green-emitting phosphor for LED applications,  
10 which makes do without special measures to stabilize it. This distinguishes it in particular from the most promising candidates to date for this purpose, namely thiogallate phosphors or chlorosilicates.

The mixed compounds with  $M = (\text{Sr}, \text{Ba})$ , preferably with up to 10% of Ba, likewise represent efficient phosphors with a wide range of emission maxima. These maxima are generally at a shorter wavelength than pure Sr Sion, preferably between 520 and 565 nm. Moreover, the color space which can be achieved can be widened by adding small amounts (preferably up to 30 mol%) of  
20 Ca and/or zinc; this shifts the emission maxima toward the longer-wave region compared to pure Sr Sion, and by partially (up to 25 mol%) replacing Si with Ge and/or Sn.

A further embodiment is for M, in particular Sr, to be partially substituted by trivalent or monovalent ions, such as  
25  $\text{La}^{3+}$  or  $\text{Li}^{+}$ . It is preferable for these ions to form at most 20 mol% of the M.

This phosphor has advantages in particular when used in an illumination system, in which case it acts as a green phosphor replacing previous inefficient solutions for the green  
30 component. The phosphor is excited either by a blue LED with



2003P14655WOUS

- 12 -

high-efficiency primary radiation or by a UV-LED. Since the green emission is in a relatively broad band compared to other technological solutions, such as thiogallates or chlorosilicates, a significantly increased color rendering index is established.

This phosphor is particularly well suited to applications in luminescence conversion LEDs which are suitable for full color and luminescence conversion LEDs with colors which can be set as desired based on an LED which primarily emits UV-blue. The conversion by the phosphor according to the invention gives blue-green to yellowish colors.

The mixed compounds with  $M = (\text{Sr}, \text{Ba})$  represent efficient phosphors with a wide range of emission maxima. These maxima are between 520 and 570 nm. Moreover, the color space which can be achieved can be widened by adding small amounts (preferably up to 30 mol%) of Ca and/or zinc and by partially (up to 25 mol%) replacing Si with Ge and/or Sn.

A further embodiment is for M, in particular Sr, to be partially substituted by trivalent or monovalent ions, such as  $\text{La}^{3+}$  or  $\text{Li}^+$ . It is preferable for these ions to form at most 20 mol% of the M.

The phosphor according to the invention can preferably be used for luminescence conversion LEDs to generate white light, albeit with blue primary radiation, but also with UV primary radiation, in which case white light is generated by means of phosphors which emit blue and yellow. Candidates for the blue component are known per se; by way of example,  $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$  (known as BAM) or  $\text{Ba}_5\text{SiO}_4(\text{Cl}, \text{Br})_6:\text{Eu}^{2+}$  or  $\text{CaLa}_2\text{S}_4:\text{Ce}^{3+}$  or also  $(\text{Sr}, \text{Ba}, \text{Ca})_3(\text{PO}_4)_3\text{Cl}:\text{Eu}^{2+}$  (known as SCAP) are suitable. The

2003P14655WOUS

- 13 -

phosphor according to the invention is suitable as the yellow component.

A red phosphor can additionally be used to improve the color of this system. It is used together with a blue-emitting base LED, and  $(Y, La, Gd, Lu)_2O_3S:Eu^{3+}$ ,  $SrS:Eu^{2+}$  or also  $(Ca, Sr)_2Si_3N_8:Eu^{2+}$ , in particular with a high Ca content, are particularly suitable.

It is in this way possible to achieve color rendering index Ra values of from 85 to 95, in wide ranges with warm-white luminous colors (emission color is not the terminus technicus?) corresponding to a color temperature from 2200 to 3000 K (??), without adversely affecting the dimmability of the illumination system.

The solution which has been discovered is therefore now superior to both sub-aspect solutions, since it allows a higher efficiency than the previous efficiency-optimized system to be achieved and also offers a much better solution to the dimmable system. The result is a breakthrough in this technology.

Particular preference is given to an illumination system in RGB technology which uses only nitride-based phosphors by using a high-efficiency blue LED with a dominant wavelength from 440 to 465 nm, preferably with a peak wavelength of 460 nm, together with luminescence conversion LEDs. A first luminescence conversion LED uses a blue LED, preferably with a peak wavelength of 460 nm, as primary light source, with conversion by means of the above-described Sr Sion as green secondary light source. A second luminescence conversion LED uses a blue LED, preferably with a peak wavelength of 460 nm, as primary light source, with conversion by means of a nitridosilicate of type  $(Ca, Sr)_2Si_3N_8:Eu^{2+}$  as red secondary light source. Surprisingly, these three components virtually ideally

2003P14655WOUS

- 14 -

complement one another in their spectrum, thereby allowing a high color rendering at high efficiency.

- The technical realization of the illumination system according to the invention can be implemented in various ways. In particular, what are known as multichip LEDs are of interest, in which the various chips (that is to say always three?) are located inside a housing. The individual chips are in this case always locally provided with the respective phosphor. For this purpose, the individual chips may be located in different cavities or together in a single cavity. The chips have in this case normally already been provided with the phosphor in a prior process. In the case of the solution with a single cavity or a solution in which the phosphor is arranged so as to be physically separate from the chip, it is also possible for the phosphor to be applied only after the chips have been installed in the housing of the illumination system. The phosphors described are particularly suitable for technologies using near-chip conversion, as are known per se from the literature, cf. for example DE 102 03 795.
- Furthermore, the invention relates to an illumination system having LEDs as described above, the illumination system also including electronic components which, by way of example, impart dimmability. A further purpose of the electronics is to actuate individual LEDs or groups of LEDs. These functions may be realized by known electronic components.

### Figures

The invention is to be explained in more detail in the text which follows on the basis of two exemplary embodiments. In the drawing:

2003P14655WOUS

- 15 -

Figure 1 shows an emission spectrum for an oxynitridosilicate;  
Figure 2 shows the reflection spectrum of this oxynitridosilicate;

5 Figure 3 shows a semiconductor component which serves as light source for white light also on a larger scale (fig. 3a);

Figure 4 shows an emission spectrum for the semiconductor component from figure 3;

10 Figure 5 shows a second exemplary embodiment of a semiconductor component.

#### Description of the drawings

Figure 1 shows a specific example for the high-efficiency, green-emitting phosphor. This example relates to the emission of the phosphor  $\text{SrSi}_2\text{N}_2\text{O}_2:(10\% \text{Eu}^{2+})$  in the HT modification, in  
15 which the Eu fraction forms 10 mol% of the lattice sites occupied by Sr. The emission maximum is at 545 nm, the mean dominant wavelength at 564 nm ( $\lambda_{\text{dom}}$ ). The color locus is  $x=0.393$ ;  $y=0.577$ . The excitation takes place at 460 nm, and the FWHM is 84 nm.

20 Figure 2 shows the diffuse reflection spectrum for this phosphor. It reveals a pronounced minimum in the range below 430 nm, which therefore demonstrates the good excitability in this range.

Figure 3 specifically illustrates the structure of a light  
25 source for white light. The light source is a semiconductor component 6 of the LED type having a first chip 1 of the InGaN type with a peak emission wavelength of, for example, 460 nm, and a second chip 2 of the InGaAlP type with a peak emission wavelength of, for example, 620 nm, and finally a semiconductor

2003P14655WOUS

- 16 -

component of the luminescence conversion LED type with a third chip 3 of the InGaN type with a primary peak emission wavelength of, for example, 460 nm. The semiconductor component 6, together with other similar elements, is embedded in an opaque basic housing 8. The phosphor is the oxynitridosilicate  $\text{SrSi}_2\text{O}_2\text{N}_2:\text{Eu}(10\%)$  proposed as the exemplary embodiment, which completely converts the primary radiation of the chip 3, transforming it into green radiation with a peak emission at 547 nm or  $\lambda_{\text{dom}} = 563$  nm. This solution has the major advantage of being tunable within a wide range of color temperatures by changing the relative intensities of the three LEDs by means of electronic control unit 7. A comparison, cf. Table 1, with the solution which has hitherto been available using three primary emitting LEDs (RGB, with green realized by an InGaN LED with  $\lambda_{\text{dom}} = 526$ ) convincingly demonstrates the superiority of the new solution. Fig. 3a shows a view of an LED on a larger scale.

Figure 4 shows the emission from an illumination system of this type as spectral distribution (intensity in arbitrary units) against the wavelength (in nm). The dashed line shows the old solution (three primary emitting LEDs) compared to the new solution (two primary emitting LEDs and a luminescence conversion LED for green) for a color temperature of 4000 K.

The particular advantage of using a long-wave primary light source (450 to 465 nm) for the green luminescence conversion LED is that this avoids problems with ageing and degradation of the housing and resin or phosphor, with the result that a long service life is achieved.

In another exemplary embodiment, a UV-LED (approximately 380 nm) is used as primary light source for the green luminescence conversion LED; in this case, problems with ageing and degradation of housing and resin or phosphor have to be

2003P14655WOUS

- 17 -

avoided as far as possible by means of additional measures which are known per se, such as careful selection of the housing material, addition of UV-resistant resin components. The major advantage of this solution is the very high efficiency of typically 30 lm/W which can thereby be achieved.

Table 1: Comparison of the color rendering index Ra and of the red index R9 between white-emitting semiconductor components based on the pure LED solution (old) and the solution using a green luminescence conversion LED (new)

Color temperature (K)	Ra (old)	R9 (old)	Ra (new)	R9 (new)
2700	38	-23	91	92
3000	38	-37	91	93
4000	43	-71	94	89
5000	36	-87	91	78
6430	51	-99	86	57

In a further exemplary embodiment, figure 5, a solution with two LEDs is used as white-emitting semiconductor component. The basic structure is similar to that described in WO 01/41215. A first luminescence conversion LED provides the blue and green components. A chip 1 of type InGaN with a primary peak emission wavelength of, for example, 460 nm is embedded in an opaque basic housing 8 in the region of a recess 9. At the same time, a second LED 2 of type InGaAlP, which emits red light, similar to the first embodiment, is also accommodated in the cavity.

2003P14655WOUS

- 18 -

The chips have separately controllable, separate terminals 3. In each case one of the terminals 3 is connected to the chip 1, 2 via a bonding wire 4. The recess has an inclined wall 7 which serves as reflector for the primary radiation of the chips 1, 2. The recess 9 is filled with a potting compound 5, which as its main constituents typically contains a silicone casting resin (or alternatively epoxy casting resin) (80 to 90% by weight) and phosphor pigments 6 (less than 15% by weight). Further small fractions can be attributed, inter alia, to methyl ether and Aerosil. The phosphor is the oxynitridosilicate  $\text{SrSi}_2\text{O}_2\text{N}_2:\text{Eu}(10\%)$  proposed as the first exemplary embodiment in a lower concentration, which only partially converts the primary radiation of the LED, transforming it into green radiation with a peak emission at 540 nm, or  $\lambda_{\text{dom}} = 560 \text{ nm}$ .

This narrow construction with a common cavity is possible since the red LED 2 with primary emission at 645 nm is not absorbed or converted by the green phosphor. This shows an example of the significance of a narrow full width at half maximum (FWHM) below 90 nm, preferably below 80 nm). The only drawback of this extremely compact solution, which has been proposed for the first time here, compared to the three LED solution is the lack of tunability.

2003P14655WOUS

- 19 -

**Claims**

1. A high-efficiency LED-based illumination system with improved color rendering, simultaneously exploiting the color-mixing principle of blue, green and red (RGB mixing) and the principle of converting a primary radiation emitted by an LED into light with a longer wavelength by means of a phosphor which absorbs this radiation, at least two LEDs being used, of which a first LED emits primarily in the range from 420 to 470 nm (dominant wavelength) and a second LED emits in the red region at 600 to 700 nm (dominant wavelength), characterized in that the green component is produced by the primary radiation of the first LED being at least partially converted by a green-emitting phosphor, the green-emitting phosphor used being a phosphor from the class of the oxynitridosilicates, having a cation M and the empirical formula  $M_{(1-c)}Si_2O_2N_2:D_c$ , M comprising Sr as a constituent and D being doped with divalent europium, with  $M = Sr$ , or  $M = Sr_{(1-x-y)}Ba_yCa_x$  with  $x+y < 0.5$  being used, the oxynitridosilicate completely or predominantly comprising the high-temperature-stable modification HT.
2. The illumination system as claimed in claim 1, characterized in that the system contains groups of LEDs of the same type.
3. The illumination system as claimed in claim 1, characterized in that three LEDs are used, the primary radiation of the first LED being completely converted into green secondary emission, with a third LED emitting



2003P14655WOUS

- 20 -

blue light unhindered, in particular in the wavelength range from 430 to 470 nm.

4. The illumination system as claimed in claim 1, characterized in that the system includes control electronics which impart dimmability.
5. The illumination system as claimed in claim 3, characterized in that the system includes control electronics which control the brightness of the individual LEDs or groups of LEDs individually, so that a tunable illumination system is formed for a range of color temperatures which covers at least 1000 K within a band from 2500 to 5000 K, with an Ra of at least 85, in particular at least 90, for each selected color temperature within the selected range.
6. The illumination system as claimed in claim 1, characterized in that two LEDs are used, the primary radiation of the first LED being only partially converted into green secondary emission, in which case the green component and the blue component are emitted simultaneously by the first LED.
7. The illumination system as claimed in claim 1, characterized in that the green secondary emission (dominant peak wavelength) is in the range from 550 to 570 nm.
8. The illumination system as claimed in claim 1, characterized in that in the oxynitridosilicate the Eu fraction makes up between 0.1 and 20 mol% of M.

2003P14655WOUS

- 21 -

9. The illumination system as claimed in claim 2, characterized in that a proportion of M, in particular up to 30 mol%, is replaced by Ba and/or Ca and/or Zn.
10. The illumination system as claimed in claim 2,  
5 characterized in that a proportion of M, in particular up to 30 mol%, is replaced by Li and/or La.
11. The illumination system as claimed in claim 2, characterized in that a proportion of SiN, in particular up to 30 mol%, is replaced by AlO.
- 10 12. The illumination system as claimed in claim 1, characterized in that a proportion of Eu, in particular up to 30 mol%, is replaced by Mn.
13. The illumination system as claimed in claim 2, characterized in that the following lines appear in its  
15 XRD spectrum:
- 
14. The illumination system as claimed in claim 2, characterized in that the following relationships between lines are present in its XRD spectrum:
- 20
- 
15. The illumination system as claimed in claim 1, characterized in that the primary radiation source used for the range from 420 to 470 nm is a light-emitting diode based on InGaN, in particular with its peak  
25 wavelength in the range from 440 to 465.
16. The illumination system as claimed in claim 1, characterized in that the color mixing using the RGB

2003P14655WOUS

- 22 -

principle realizes a white-emitting illumination system with a color temperature of from 2500 to 5000 K, in particular 3500 to 5000 K.

17. The illumination system as claimed in claim 1,  
5 characterized in that a plurality of light-emitting components, in particular a luminescence conversion LED which imparts blue and green components of the emission simultaneously and an LED which imparts red components of the emission, are arranged in a cavity.
- 10 18. The illumination system as claimed in claim 17, characterized in that the full width at half maximum of the oxynitridosilicate is less than 90 nm, preferably less than 80 nm.
- 15 19. The illumination system as claimed in claim 1, characterized in that the system includes electronics for actuating individual LEDs or groups of LEDs.

2003P14655WOUS

- 23 -

**Abstract****High-efficiency LED-based illumination system with improved color rendering**

The illumination system makes simultaneous use of the color-mixing principle from blue, green and red (RGB mixing) and the principle of converting of a primary radiation emitted by an LED into light with a longer wavelength by a phosphor which absorbs this radiation, with at least two LEDs being used, of which a first LED emits primarily in the range from 420 to 470 nm (dominant wavelength) and a second LED emits in the red region at 600 to 700 nm (dominant wavelength), wherein the green component is produced by the primary radiation of the first LED being at least partially converted by a green-emitting phosphor, the green-emitting phosphor used being a phosphor from the class of the oxynitridosilicates, having a cation M and the empirical formula  $M_{(1-c)}Si_2O_2N_2:D_c$ , M comprising Sr as a constituent and D being doped with divalent europium, where  $M = Sr$  or  $M = Sr_{(1-x-y)}Ba_yCa_x$  with  $x+y < 0.5$  is used, the oxynitridosilicate completely or predominantly comprising the high-temperature-stable modification HT.

Fig. 1

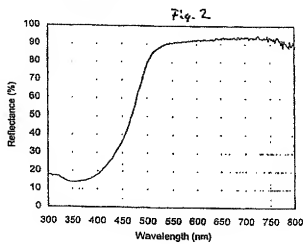
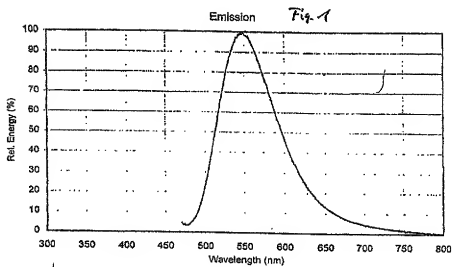




Fig. 3a

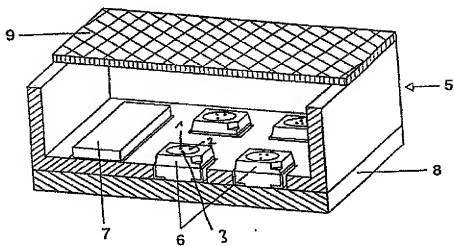


FIG. 3

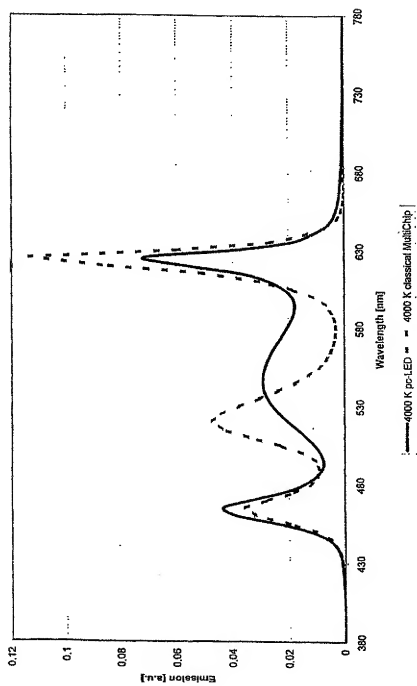


Fig. 4

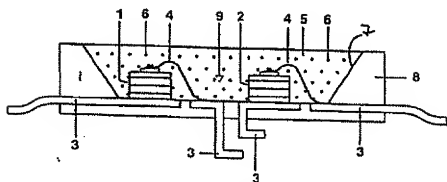


FIG. 5



# EXHIBIT 2

Leuchtstoff-Type: LED-Leuchtstoff

Datum: 07.05.03

Veranlassung/ grünes Sr-SiON

Ziel:  $\text{Sr}_{0,99}\text{Eu}_{0,01}\text{Si}_2\text{O}_7\text{N}_2$

Versuchsbeschreibung (Ansatz, Bearbeitung, usw.):

Substanz	M ( $^{100}\text{mol}$ )	Element	El-Masse	Atomanteil	$I_1$ -Einwaage	$I_2$ -Einwaage
$\text{SiO}_2$ Aerosil 90	60,090	Si	60,090	0,500	30,045 g	2,003 g
$\text{Si}_3\text{N}_4$ UBE-SN10	140,283	Si	46,761	1,500	70,142 g	4,676 g
$\text{SrCO}_3$ Sphäral	147,630	Sr	147,630	0,950	140,249 g	9,350 g
$\text{SrF}_2$ Rohf	125,620	Sr	125,620	0,003	0,377 g	0,025 g
$\text{Eu}_2\text{O}_3$ 99,99% RP	351,920	Eu	175,960	0,050	8,798 g	0,587 g
Gesamteinwaage					219,57	16,641 g

Bei diesem Versuch wurde  $\text{SrF}_2$  als Schmelzmittel eingesetzt.

Die Ausgangssubstanzen wurden an Luft in eine 100mL PE-Flasche eingewogen und in i-Propanol mit 5 kleinen Achatkugeln in der dichtverschlossenen PE-Flasche 4 Stunden auf dem Rollbock homogenisiert. Die Mischung wurde in einen mit W-Blech ausgekleideten Mo-Schalentiegel gefüllt und mit Deckel bei  $1540^\circ\text{C}$  8 Stunden lang (Aufheizrate:  $250\text{K/h}$ , Abkühlrate:  $130\text{K/h}$ ) geglüht. Das Rohr wurde während der Glühung mit  $200\text{L/h}$  7,5%-Formiergas gespült.

Die gesamte Probe besitzt eine sehr saubere gelb-grüne Körperfarbe.

Das Material wird 5min bzw. 10min in der Mörsermühle gemahlen und über einer  $54\mu\text{m}$  Gase gesiebt.

Bearbeitung: 1. Glühung

Glühofen	GERO-RO
Glühtemperatur ( $^\circ\text{C}$ )	1540 $^\circ\text{C}$
Glühatmosferaere	FG
$I_1$	200 <small>N<math>^\circ</math> 90 (h) FG, 150 (h)</small>
Glühdauer (h)	8,0
Schalenmaterial	W-Tiegel
Füllmengen (g)	15,0

Ergebnis:

Konsistenz	nicht gesintert
Körperfarbe	gelb-grün
Lumineszenz	
Anregung	
Remission	
Röntgen	

# EXHIBIT 3

## Phosphor test report

Test No.:  
TF 119/2003**Phosphor type:** LED phosphor

Date: 05.07.03

**Cause/objective:** green Sr-SiON  
 $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{O}_7\text{N}_2$ **Test description (approach, process, etc.):**

Substance	$M(^{\circ}/\text{mol})$	Element	El-Mass	Atomic content	$^{1/1}$ Weighed content	$^{1/33}$ Weighed content
$\text{SiO}_2$ , Aerosil 90	60.090	Si	60.090	0.500	30.045 g	2.003 g
$\text{Si}_3\text{N}_4$ , UBE-SN10	140.283	Si	46.761	1.500	70.142 g	4.676 g
$\text{SrCO}_3$ , Solvay	147.630	Sr	147.630	0.950	140.249 g	9.350 g
$\text{SrF}_2$ , RdH	125.620	Sr	125.620	0.003	0.377 g	0.025 g
$\text{Eu}_2\text{O}_3$ , 99.99%, RP	351.920	Eu	175.960	0.050	8.798 g	0.587 g
<b>Total weighed content</b>					<b>219.57</b>	<b>16.641 g</b>

The flux used in this test was  $\text{SrF}_2$ .

The starting substances were weighed into a 100 ml PE bottle under air and homogenized in i-propanol using 5 small rolling agate beads for 4 hours in the sealed PE bottle. The mixture was transferred into a Mo dish crucible lined with a W metal plate and was then calcined for 8 hours at 1540°C with a lid (heat-up rate: 250 K/h, cooling rate: 130 K/h). The tube was purged with 200 l/h of 7.5% forming gas during the calcining.

The entire specimen has a very clean yellow-green body color.

The material is milled in a pestle mill for 5 or 10 min and screened over a 54 µm gauze.

**Processing:** 1st calcining

Calcining furnace: GERO-RO

Calcining temperature (°C): 1540°C

Calcining atmosphere: FG

 $t_h$ : 200 N2: 50 l/h  
PG: 150 l/h

Calcining duration (h): 8.0

Dish material: W crucible

Fill quantities (g): 15.0

**Results:**

Consistency

not sintered

Body color

yellow-green

Luminescence

Excitation

Reflectance

X-ray

# EXHIBIT 4

Leuchtstoff-Type: **LED-Leuchtstoff**

Datum: 07.05.03

Veranlassung/ **grünes Sr-SION**Ziel:  **$\text{Sr}_{0.99}\text{Eu}_{0.05}\text{Si}_2\text{O}_7\text{N}_2$** Versuchsbeschreibung (Ansatz, Bearbeitung, usw.):

Substanz	M ( $^{100}_{\text{mol}}$ )	Element	El-Masse	Atomanteil	$I_1$ -Einwaage/ $_{35}$ -Einwaage
$\text{SiO}_2$ Aerosil 90	60.090	Si	60.090	0.500	30.045 g 2.003 g
$\text{Si}_3\text{N}_4$ UBE-SN10	140.283	Si	46.761	1.500	70.142 g 4.676 g
$\text{SrCO}_3$ Schvay	147.630	Sr	147.630	0.950	140.249 g 9.350 g
$\text{SrF}_2$ RdH	125.620	Sr	125.620	0.003	0.377 g 0.025 g
$\text{Eu}_2\text{O}_3$ 99.99%, R+P	351.920	Eu	175.960	0.050	8.798 g 0.587 g
Gesamteinwaage					219.57 16.641 g

Bei diesem Versuch wurde  $\text{SrF}_2$  als Schmelzmittel eingesetzt.

Die Ausgangssubstanzen wurden an Luft in eine 100mL PE-Flasche eingewogen und in i-Propanol mit 5 kleinen Achatkugeln in der dichtverschlossenen PE-Flasche 4 Stunden auf dem Rollbock homogenisiert.

Die Mischung wurde in einen mit W-Blech ausgekleideten Mo-Schalentiegel gefüllt und mit Deckel bei  $1540^\circ\text{C}$  8 Stunden lang (Aufheizrate:  $250\text{K/h}$ , Abkühlrate:  $130\text{K/h}$ ) geglüht. Das Rohr wurde während der Glühung mit  $200\text{ L/h}$  7,5%-Formiergas gespült.

Die gesamte Probe besitzt eine sehr saubere gelb-grüne Körperfarbe.

Das Material wird 5min bzw. 10min in der Mörsermühle gemahlen und über einer  $54\mu\text{m}$  Gase gesiebt.

Bearbeitung: **1. Glühung**Glühofen GERO-ROGlühtemperatur ( $^\circ\text{C}$ ) 1540Glühatmosphäre FG $I_h$  200 NO 100 h  
PG 100 hGlühdauer (h) 8.0Schalenmaterial W-TiegelFüllmengen (g) 15.0Ergebnis:

Konsistenz nicht gesintert  
Körperfarbe gelb-grün  
Lumineszenz 


  
Anregung 


  
Remission 


  
Röntgen 


## Leuchtstoffversuchsprotokoll

Versuchs-Nr.:

**TF 163/2003**

Datum: 07.05.03

**Leuchtstoff-Type:**

## LED-Leuchtstoff

**Veranlassung/**

### grünes Sr-SION

**Ziel:**

$$\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{O}_2\text{N}_2$$

**Versuchsbeschreibung (Ansatz, Bearbeitung, usw.):**

Bei dieser Probe handelt es sich um die 10 min gemahlene TF 119/03. Die Probe wurde unter der neuen Bezeichnung TF163/03 an OS zur LED-Anprobe gegeben.

**Bearbeitung:**

## 1. Glühung

## Glühofen

GERO-RO

Glühtemperatur (°C)

1540°C

## Glühatmosphäre

FG

1/2

200

N2 50 1m

Glühdauer (h)

8.0

### Schalenmaterial

W-Tiegel

Füllmengen (g)

15.0

**Ergebnis:**

### Konsistenz

nicht gesintert

Körperfarbe

gelb-grün

## Lumineszenz

### Anregung

## Remission

## Röntgen

Leuchtstoff-Type:**LED-Leuchtstoff**

Datum: 07.05.03

Veranlassung/grünes Sr-SiONZiel: $\text{Sr}_{0.99}\text{Eu}_{0.01}\text{Si}_2\text{O}_7\text{N}_2$ Versuchsbeschreibung (Ansatz, Bearbeitung, usw.):

Bei dieser Probe handelt es sich um die 5 min gemahlene TF 119/03. Die Probe wurde unter der neuen Bezeichnung TF164/03 an OS zur LED-Anprobe gegeben.

Bearbeitung:**1. Glühung**

Glühofen

GERO-RO

Glühtemperatur (°C)

1540°C

Glühatmosphäre

FG

 $I_{th}$ 

200

N2 200 h

F-G 100 h

Glühdauer (h)

8.0

Schalenmaterial

W-Tiegel

Füllmengen (g)

15.0

Ergebnis:

Konsistenz

nicht gesintert

Körperfarbe

gelb-grün

Lumineszenz

Anregung

Remission

Röntgen



# EXHIBIT 5

**Phosphor type:** LED phosphor

Date: 05.07.03

**Cause/objective:** green Sr-SiON  
 $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{O}_7\text{N}_2$ **Test description (approach, process, etc.):**

Substance	$M(^{\circ}\text{mol})$	Element	El-Mass	Atomic content	$1/1$ Weighed content	$1/31$ Weighed content
$\text{SiO}_2$ , Aerosil 90	60.090	Si	60.090	0.500	30.045 g	2.003 g
$\text{Si}_3\text{N}_4$ , UBE-SN10	140.283	Si	46.761	1.500	70.142 g	4.676 g
$\text{SrCO}_3$ , Solvay	147.630	Sr	147.630	0.950	140.249 g	9.350 g
$\text{SrF}_2$ , ReH	125.620	Sr	125.620	0.003	0.377 g	0.025 g
$\text{Eu}_2\text{O}_3$ , 99.99%, RF	351.920	Eu	175.960	0.050	8.798 g	0.587 g
<b>Total weighed content</b>					219.57	16.641 g

The flux used in this test was  $\text{SrF}_2$ .

The starting substances were weighed into a 100 ml PE bottle under air and homogenized in i-propanol using 5 small rolling agate beads for 4 hours in the sealed PE bottle. The mixture was transferred into a Mo dish crucible lined with a W metal plate and was then calcined for 8 hours at 1540°C with a lid (heat-up rate: 250 K/h, cooling rate: 130 K/h). The tube was purged with 200 l/h of 7.5% forming gas during the calcining.

The entire specimen has a very clean yellow-green body color.

The material is milled in a pestle mill for 5 or 10 min and screened over a 54 µm gauze.

**Processing:** 1st calcining

Calcining furnace

GERO-RO

Calcining temperature (°C)

1540°C

Calcining atmosphere

FG

 $1/1$ 

200

N2: 50 l/h

FG: 150 l/h

Calcining duration (h)

8.0

Dish material

W crucible

Fill quantities (g)

15.0

**Results:**

Consistency

not sintered

Body color

yellow-green

Luminescence

Excitation

Reflectance

X-ray

**Phosphor type:** LED phosphor

Date: 05.07.03

**Cause/objective:** green Sr-SiON  
 $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{O}_7\text{N}_2$ **Test description (approach, process, etc.):**

This specimen is the TF 119/03 which has been milled for 10 mln. The specimen, under the new name TF 163/03, was passed to OS for LED testing.

**Processing:** 1st calcining

Calcining furnace GERO-RO

Calcining temperature (°C) 1540°C

Calcining atmosphere FG

 $t_h$  200
 

N2: 50 l/h  
 PG: 150 l/h

Calcining duration (h) 8.0

Dish material W crucible

Fill quantities (g) 15.0

**Results:**

Consistency

not sintered

Body color

yellow-green

Luminescence

Excitation

Reflectance

X-ray


OSRAM

DP-M

## Phosphor test report

Test No.:

TF 164/2003

**Phosphor type:** LED phosphor

Date: 05.07.03

**Cause/objective:** green Sr-SiON  
 $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{O}_7\text{N}_2$ **Test description (approach, process, etc.):**

This specimen is the TF 119/03 which has been milled for 5 min. The specimen, under the new name TF 164/03, was passed to OS for LED testing.

**Processing:** 1st calcining

Calcining furnace GERO-RO

Calcining temperature (°C) 1540°C

Calcining atmosphere FG

 $\frac{1}{h}$  200
 

RZ: 50 1/h  
 FG: 150 1/h

Calcining duration (h) 8.0

Dish material W crucible

Fill quantities (g) 15.0

**Results:**

Consistency

not sintered

Body color

yellow-green

Luminescence

Excitation

Reflectance

X-ray

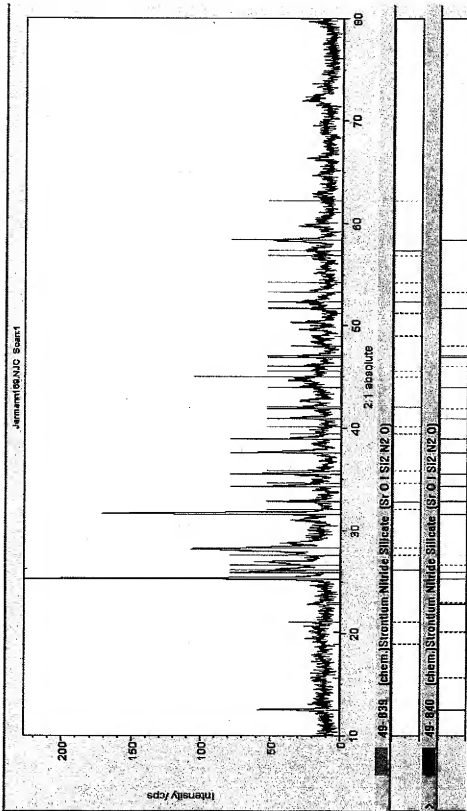

# EXHIBIT 6

SEE THE WORLD IN A NEW LIGHT

OSRAM

Schrittweite [°]: 0.01

Winkelbereich [°2 $\theta$ ]: 10-80  
Schrittweite [°]: 0.01  
Maßzeit/Schritt [s]: 1  
Probe: TF 168/03  $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{N}_2\text{O}_2$   $\text{SrCO}_3$  Solvay,  $\text{SiO}_2$  A90, 8h/1520°C  
I-Propanol gemischt, 20 min. gemahlen

Probe: TF 168/03  $\text{Sr}_{0.95}\text{Eu}_{0.05}\text{Si}_2\text{N}_2\text{O}_2$ 

**Röntgenlfraktometer Selfert URD 6**  
**Operator: C. Koch**

17.07.2003

# **EXHIBIT 7**

DMC-SM

SEE THE WORLD IN A NEW LIGHT



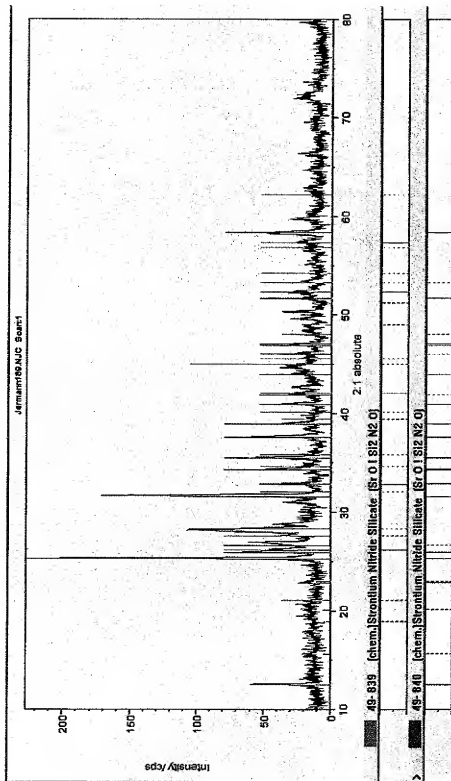
OSRAM

Angle range [ $2\theta$ ]: 10-80

Increment [ $^\circ$ ]: 0.01

Measurement time/step [s]: 1

Specimen: TF 168/03  $\text{Sr}_{0.98}\text{Eu}_{0.02}\text{Si}_3\text{N}_8\text{O}_2$   $\text{SrCO}_3$  Solvay,  $\text{SiO}_2$  A90, 8h/1520 $^\circ\text{C}$  mixed with I-propanol, milled for 20 min



X-ray diffractometer Seifert URD 6  
Operator: C. Koch

07.17.2003



# EXHIBIT 8

Vertraulich!

An OSRAM GmbH  
Abteilung: PAT-M

**ERFINDUNGSMELDUNG**  
Bitte verschlossen weitersenden!

Aktenzeichen der PAT

2003 E 12122 DE

Ihr/Wir (vor- und Nachname des/dar Erfinders/-gen - weitere Angaben und Unterschriften) letzte Seite  
Tim Fiedler, Frank Jermann, Martin Zachau, Herbert Brunner

Datum der Abfertigung:

melde(n) hiermit die auf den folgenden Seiten vollständig beschriebene Erfindung mit der Bezeichnung:  
Durchstimmbare Dreh-Chip RGB-LED mit verbesserter Farbwiedergabe

<p>I. An Vorgesetzten des/des Erfinders[en]</p> <p>Herrn/Frau Dr. Klam R&amp;D-M (Dienststelle)</p> <p>mit der Bitte, die nachstehenden Fragen zu beantworten:</p> <p>a) Wann ging die Erfindungsmeldung bei Ihnen ein? b) Geht die Erfindung auf öffentlich geförderte Arbeiten zurück? <input checked="" type="checkbox"/> nein <input type="checkbox"/> ja, Projekt (Vorhaben): c) Bitte bei Zuständigkeit auch zu Ziffer II. Stellung nehmen.</p> <p>Datum (Unterschrift des Vorgesetzten)</p>	<p>Eingang am:</p> <p>Ab Eingang laut gesetzliche Frist</p>
<p>II. Bitte wegen gesetzlicher Frist sofort weiterleiten an PAT-M zur weiteren Veranlassung.</p>	<p>GC P-M Eingang Eingang am: 1. Aug. 2003 OSRAM Frist:</p>
<p>III. An Bereichs- bzw. Werksleitung</p> <p>Herrn/Frau Dr. Klam R&amp;D-M (Dienststelle)</p> <p>Zur Entscheidung bzw. Empfehlung:</p> <p><input type="checkbox"/> Die Erfindung sollte unbeschränkt in Anspruch genommen werden. <input type="checkbox"/> Anmeldung wird empfohlen. <input type="checkbox"/> Die Erfindung kommt für eine Behandlung als Betriebsgeheimnis in Betracht. <input type="checkbox"/> Die Erfindung kommt evtl. für Auslandsanmeldungen in Betracht. Länder: <input type="checkbox"/> Die Erfindung sollte freigegeben werden, da für den Betrieb wirtschaftliche Verwertung nicht ersichtlich. <input type="checkbox"/> Bei Freigabe sollte ein Mitbenutzungsrecht gesichert werden. <input type="checkbox"/> Die Freigabe kann vorbehaltlos erfolgen, d.h. auf ein Mitbenutzungsrecht kann verzichtet werden. <input type="checkbox"/> Vor Freigabe sind vorsorglich noch folgende Dienststellen zu befragen:</p> <p>Dringlichkeitsvermerk</p> <p>(Gefüge) (Unterschrift/des Bereichs- bzw. Werksleitung)</p>	<p>Eingang am:</p> <p>Vermerke der PAT-M</p>
<p>IV. Zurückarbeiten an PAT-M</p>	

1. Welches technische Problem soll durch Ihre Erfindung gelöst werden?
2. Wie wurde dieses Problem bisher gelöst?
3. In welcher Weise löst Ihre Erfindung das angegebene technische Problem?

1. Für bestimmte Anwendungen ist der Einsatz von RGB-LEDs interessant, die aus drei Chips mit den Emissionsfarben Rot, Grün und Blau bestehen. Da alle drei Farben unabhängig voneinander angesteuert werden können, ist es mit dieser Art von LED möglich, nahezu jeden gewünschten Farbtönen über eine entsprechende Steuerelektronik gezielt einzustellen. Ein Nachteil dieser Lösung ist eine sehr niedriger Farbwiedergabeindex  $R_{fr} < 50$ , der sich aufgrund der Schmalbandigkeit der drei Einzelmissionen ergibt. Ein weiterer Nachteil ist, dass die eingesetzte Grün-LED aus technologischen Gründen deutlich weniger effizient ist als die rote und die blaue LED. In GaN-Chips sind im grünen Emissionsbereich bereits deutlich weniger effizient und der Farbtönen stark Strom und Temperaturabhängig. In GaAlP-Chips sind im grünen Wellenlängenbereich nicht sehr effizient und erst ab Emissionswellenlängen von ca. 540 nm Wellenlänge sinnvoll herstellbar.

2. Für Anwendungen, bei denen die Durchstimmbarkeit der Emissionsfarbe wichtig ist, wird die oben beschriebene LED angeboten. Für Anwendungen mit höheren Anforderungen an die Farbwiedergabe werden LEDs angeboten, bei denen ein Teil der LED-Emission über Konversion mit einem oder mehreren Leuchtstoffen generiert wird. Diese Art von LEDs ist jedoch nicht durchstimmbar.

3. Mit dem neuen grünen Leuchtstoff  $\text{SrSiO}_2\text{N}_2\text{:Eu}$  kann die relativ ineffiziente Grün-LED durch eine effizientere Konversions-LED (Basis Blau-LED + Sr-SiON) ersetzt werden. Da die grüne Emission zudem breitbandiger ist, ist mit einem deutlich gestiegenen Farbwiedergabeindex zu rechnen. Ein weiterer Vorteil ist die höhere Farbstabilität der grünen Konversions-LED. In einer erweiterten Variante könnte auch die rote LED durch eine rote Konversions-LED (Basis: Blau-LED + Roteleuchtstoff, z. B.  $(\text{Sr,Ca})_2\text{SiO}_3\text{N}_2\text{:Eu}$ ) ersetzt werden. Auf diese Art und Weise ließen sich in weiten Bereichen bei warmweißen Emissionsfarben  $R_{fr}$ -Werte zwischen 85 und 95 erzielen. Man hätte also eine voll durchstimmbare LED mit guter Farbwiedergabe erreicht. Gegenüber 1-Chip-Konversions-LEDs ergibt sich als weiterer Vorteil eine höhere Effizienz, da die Reabsorption von grüner Lumineszenzstrahlung durch den roten Leuchtstoff vermieden wird.

Beide Lösungen sind möglich geworden, durch die Entwicklung effizienter Nitrid-Leuchtstoffe, die deutlich stabiler sind als die bisher bekannten Sulfidleuchtstoffe (grünes Thioallat:  $\text{Eu}$ , rotes  $(\text{Sr,Ca})\text{S:Eu}$ ) und sich überraschenderweise im Zusammenspiel mit der blauen 460 nm-LED nahezu ideal in ihrem Spektrum ergänzen. Insbesondere sind sog. Multichip-LEDs von Interesse. Hierbei befinden sich die Chips in einem Gehäuse. Die einzelnen Chips sind dabei lokal mit dem jeweiligen Leuchtstoff versehen. Die Chips können sich zu diesem Zweck in verschiedenen Kavitäten befinden oder in einer. Bei der letzteren Variante können die Chips mit dem benötigten Leuchtstoff bereits in einem Vorprozess versehen werden oder nach dem Einbau in das Gehäuse. Alle Patente von OSRAM zur chipnahen Konversion insbesondere mit sog. Dünnfilm-Chips sind hier zu erwähnen.

\* oder UV-LED (siehe „Pure Green“ Erfindungsquelle)

Fr 08.08.08

4. Zur weiteren Erläuterung sind als Anlagen beigelegt:

- \_\_\_\_\_ Blatt der Darstellung eines oder mehrerer Ausführungsbeispiele der Erfindung;
- \_\_\_\_\_ Blatt zusätzliche Beschreibungen (z.B. Laborberichte, Versuchsprotokolle);
- \_\_\_\_\_ Blatt Literatur, die den Stand der Technik, von dem die Erfindung ausgeht, beschreibt;
- \_\_\_\_\_ Blatt sonstige Unterlagen;

Confidential information contained in this page is redacted.

# EXHIBIT 9

<b>Confidential!</b> To OSRAM GmbH Department: GC IP-M	<b>NOTIFICATION OF INVENTION</b> <b>Please forward under sealed cover!</b>	GC IP reference 2003E12122DE
I/we (full name of the inventor[s] - further details and signature[s] on last page) Tim Fiedler, Frank Jermann, Martin Zachau, Herbert Brunner		Date of signature:
hereby notify you of the invention which is described in full on the following pages, named:  Tunable three-chip RGB-LED with improved color rendering		
<b>I.</b>	<b>To the responsible PLA, with a request for immediate forwarding to GC IP</b>  Mr/Ms <u>Dr. Klam</u> <span style="float: right;">R&amp;D-M</span> <small>(Section)</small> Please answer the following questions: a) When did you receive the notification of invention? b) Is the invention derived from publicly funded work? <input checked="" type="checkbox"/> no <input type="checkbox"/> yes, project (planned): _____ c) If responsible, please also offer opinion on section III. <input type="checkbox"/>  Date _____ <small>(Signature of the PLA)</small>	Received on:  9.1.03     Statutory time limit runs from date of receipt!
<b>II.</b>	<b>Because of the statutory time limits please forward immediately to GC IP-M for further processing.</b>	Received at GC IP-M on: Sept. 1, 2003 OSRAM time limit:
<b>III.</b>	<b>To the OPT</b>  <u>Dr. Klam</u> <span style="float: right;">R&amp;D-M</span> <small>(Section)</small> For decision:  <input type="checkbox"/> The invention should be used without restriction. <input type="checkbox"/> Application recommended.  <input type="checkbox"/> The invention can be considered for treatment as a trade secret.  <input type="checkbox"/> The invention can be considered for foreign applications. Countries: _____  <input type="checkbox"/> The invention should be released as there is no apparent economic utilization for the company. <input type="checkbox"/> On release, a right of joint usage should be secured.  <input type="checkbox"/> The release can take place unconditionally, i.e. a right of joint usage is not required.  <input type="checkbox"/> Before release, the following sections should be questioned as a precaution: _____  <div style="display: flex; justify-content: space-between; align-items: flex-end;"> <div data-bbox="142 1185 407 1221">           9.1.03    <u>[signature]</u>  <small>(Date)                      (Cost center manager's signature)</small> </div> <div data-bbox="583 1148 723 1164" style="border: 1px solid black; padding: 2px;"> <b>Notes as to urgency</b> </div> </div>	Received on:  9.1.03     <div style="border: 1px solid black; padding: 5px; margin-top: 10px;"> <b>Notes by GC IP-M</b>          First copy of the electronic notification of 7.29 was already with OG on 8.12.03       </div>
<b>IV.</b>	<b>Please return to GC IP-M    Km → We</b>	

1. What technical problem is your invention supposed to solve?
2. How was this problem previously solved?
3. How does your invention solve the stated technical problem?

1. The use of RGB-LEDs, which consist of three chips with red, green and blue emission colors, is of interest for certain applications. Since all three colors can be actuated independently of one another, this type of LED makes it possible to deliberately set virtually any desired color locus by means of correspondent control electronics. One drawback of this solution is a very low color rendering index  $R_a < 50$  which results from the narrow-band nature of the three individual emissions. A further drawback is that the green LED used, for technological reasons is significantly less efficient than the red and blue LEDs. In the green emission region, InGaIn chips are already significantly less efficient and the color locus is highly current and temperature dependent. InGaAlP chips are not very efficient in the green wavelength region and can only be economically produced beyond emission wavelengths of approx. 540 nm.

2. The LED described above is recommended for applications where the tunability of the emission color is important. LEDs in which part of the LED emission is generated by conversion with one or more phosphors are recommended for applications with higher color rendering requirements. However, this type of LEDs is not tunable.

3. By using the new green phosphor  $\text{SrSi}_2\text{O}_2\text{N}_2:\text{Eu}$ , the relatively inefficient green LED can be replaced with a more efficient conversion LED (blue base LED\* + Sr-SiON). Since, moreover, the green emission has a wider band, a considerably increased color rendering index is likely. A further advantage is the higher color locus stability of the green conversion LED.

In a wider variant, the red LED could also be replaced with a red conversion LED (blue base LED + red phosphor, e.g.  $(\text{Sr},\text{Ca})_2\text{SiS}_8:\text{Eu}$ ). In this way,  $R_a$  values of between 85 and 95 can be achieved over wide regions with warm-white emission colors. This would give a fully tunable LED with good color rendering. A further advantage over 1-chip conversion LEDs is a higher efficiency, since the reabsorption of green luminescence radiation by the red phosphor can be avoided.

Both solutions are made possible by the development of efficient nitride phosphors, which are considerably more stable than the known sulphide phosphors (green thiogallate:Eu, red  $(\text{Sr},\text{Ca})_2\text{S}:\text{Eu}$ ) and whose spectrum surprisingly forms a virtually ideal complement to the blue 460 nm LED. So-called multichip LEDs are of particular interest. Here, the chips are located inside one housing. The individual chips are locally provided with the respective phosphor. For this purpose, the chips may be located in different cavities or in one cavity. In the latter variant, the chips may be provided with the required phosphor during a preliminary process or after they have been fitted into the housing. All OS/OSRAM patents relating to near-chip conversion, in particular using thin-film chips, should be mentioned here.

\* or UV-LED (see "Pure Green" notification of the invention) [FJ] 08.06.03

4. By way of additional explanation, the following are enclosed as annexes:

\_\_\_\_\_ pages setting out one or more exemplary embodiments of the invention;  
\_\_\_\_\_ pages of additional descriptions (e.g. laboratory reports, experimental records);  
\_\_\_\_\_ pages of literature describing the prior art on which the invention is based;  
\_\_\_\_\_ pages of other documents:

5. Which sections are interested in the invention? OSRAM OS
6. Has the invention already been tested (experiments carried out, samples prepared)?  
☒ no ☐ yes, result: \_\_\_\_\_
7. For which products is the invention applicable? True-color RGB LED
8. Is the use of the invention planned?  
☐ no ☒ yes, by: OSRAM OS
9. Is a product supplied which is based on the invention or is it intended that such a product be supplied?  
☒ no ☐ yes, (probably) on \_\_\_\_\_; name of the product: \_\_\_\_\_
10. Is it intended that the invention be published or has this already occurred?  
☒ no ☐ yes (probably) on \_\_\_\_\_ in the following book, magazine: \_\_\_\_\_
11. Is it intended that the invention be notified to persons outside the company or has this already occurred?  
☒ no ☐ yes (probably) on \_\_\_\_\_ to \_\_\_\_\_
12. Personal details of the inventor[s] (Enter inventors 1 - 4 here. Attach additional sheet for further inventors):

Surname	Fiedler	Jermann	Zachau	Brunner
First name	Tim	Frank	Martin	Herbert
Academic level/title/profession	Dipl.-Ing.	Dr. rer. Nat.	Dr. rer. Nat.	Dipl.-Ing.
Section and site	DP-M	DP-M	DP-M	OS T BT1
Activity/position in the company (e.g. project manager etc.)	Technician	Technician	Head of Department	Head of Department
Telephone extension	3228	3357	3366	1209
Nationality	German	German	German	German
Private address with post code	81543 Munich	81739 Munich	82269 Geltendorf	93047 Regensburg
and street, house No.	Hans-Mielich-Str. 7	Klara-Ziegler-Bogen 187	Pfarrer-Unsinn-Str. 17	Winklergasse 16
Date of birth	02.25.76	11.02.1966	12.23.1960	05.18.64
Accounting personnel office or PA No.	PA-M	PA-M	PA-M	PA-OS
Personnel number *)	749-3001	749-2899	Zachau	00618665
13. Is the invention within a) your area of work? b) one of your employer's other areas of work?	<input checked="" type="checkbox"/> yes <input type="checkbox"/> no <input checked="" type="checkbox"/> yes <input type="checkbox"/> no	<input checked="" type="checkbox"/> yes <input type="checkbox"/> no <input checked="" type="checkbox"/> yes <input type="checkbox"/> no	<input checked="" type="checkbox"/> yes <input type="checkbox"/> no <input checked="" type="checkbox"/> yes <input type="checkbox"/> no	<input checked="" type="checkbox"/> yes <input type="checkbox"/> no <input checked="" type="checkbox"/> yes <input type="checkbox"/> no
14. What is your share of the invention?	30%	30%	20%	20%
15. Has the invention also been notified, or is it also being notified, as an improvement suggestion?	<input type="checkbox"/> yes <input checked="" type="checkbox"/> no	<input type="checkbox"/> yes <input checked="" type="checkbox"/> no	<input type="checkbox"/> yes <input checked="" type="checkbox"/> no	<input type="checkbox"/> yes <input checked="" type="checkbox"/> no
16. If you regard the invention as an independent invention, please give reasons:				
17. No other persons are, to my/our knowledge, involved in the invention.	T. Fiedler 08.06.03 (Signature)	Frank Jermann 08.06.03 (Signature)	[signature] 8.8.03 (Signature)	7.29.03 [signature] (Signature)



# EXHIBIT 10

**OSRAM**

Epping, Herrmann Fischer  
Patentanwalts-gesellschaft mbH  
Herrn Dipl.-Phys. Roland Schachtner  
Ridlerstraße 55

80339 München

Name: Susanne Kämmerle  
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Ihr Schreiben: 2003E12122DE  
Unser Zeichen: KM  
Datum: 05.09.2003

**Erfindungsmeldung**  
**Titel:**

2003E12122DE  
Durchstimmbare Drei-chip RGB-LED mit verbesserter  
Farbwiedergabe

**Erfinder:**

Fiedler – OG, Dr. Jermann – OG, Dr. Zachau – OG, Brunner –  
OOS

Inanspruchnahme ohne, Inc.:

Alle OG-Erfinder, abgesandt durch OG: 04.09.03

Sehr geehrter Herr Schachtner,

In der Anlage erhalten Sie eine Kopie der oben genannte Erfindungsmeldung.  
Bitte wickeln Sie die Inanspruchnahme für die OOS-Erfinder in gewohnter Weise ab.

Die weitere Bearbeitung wird Herr Dr. Wemar übernehmen.  
Es ist eine Gemeinschaftsanmeldung OOS/PTG vorgesehen. Sobald die Erstanmeldung erfolgt ist,  
werden wir wieder berichten.

Mit freundlichen Grüßen

  
Susanne Kämmerle

Anlage: Erfindungsmeldung

Kopie (mit Anl.): Dr. Nirschl, OOS-R.

Belästigung:  
OSRAM GmbH  
D-81235 München

Hausanschrift:  
OSRAM GmbH  
Hofbrunngraben Straße 1  
D-81543 München

OSRAM-Gesellschaft  
mit beschränkter Haftung  
München  
Vorstandsvorsitzender  
Aufsichtsrats:  
Jürgen Radonski

Geschäftsführung:  
Wolfgang Bapst,  
Vorstandsvorsitzender  
Thomas Seeburg  
Jörg Scheller

Registrierungs-  
München, HRB 4151

# EXHIBIT 11

[Letter from Osram to Epping dated 09.05.2003]

Notification of invention    2003E12122DE  
Title:                            Tunable three-chip RGB-LED with  
                                 improved color rendering  
Inventors:                       Fiedler - OG, Dr. Jermann - OG,  
                                 Dr. Zachau - OG, Brunner - OOS  
Claim without Inc.:           All OG inventors, sent by OG:  
                                 09.04.03

Dear Mr Schachtner,

Please find enclosed a copy of the abovementioned notification of invention. Please handle the claim of the OOS inventors in the usual way.

Dr. Werner will be responsible for further processing. A joint OOS/PTG application is planned. Once the first application has been made, we will report back.

Yours sincerely

[signature]

Susanne Kärmerle

Enc.: Notification of invention

Copy (with enc.): Dr. Nirschl, OOS-R